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ANTIMICROBIENNE

(54) MULTIPHASE CLEANER WITH ANTIMICROBIAL ACTION

(57) A liquid multiphase cleaner having at least two continuous phases, which has at least one aqueous phase I and a nonaqueous liquid phase II which is immiscible with this aqueous phase, and which can be converted temporarily into an emulsion by shaking, comprises at least one antimicrobial active ingredient, together with a spray dispenser forms a product and can be used in a method for the cleaning and/or disinfection or sanitation of hard surfaces, in particular glass, in which the liquid multiphase cleaner is converted temporarily into an emulsion by shaking, is applied to the surface to be cleaned and/or disinfected or sanitized, preferably by spraying, and this surface is then cleaned and/or disinfected or sanitized, if necessary by wiping with an absorbent soft article.

ABSTRACT

5 A liquid multiphase cleaner having at least two continuous
phases, which has at least one aqueous phase I and a
nonaqueous liquid phase II which is immiscible with this
aqueous phase, and which can be converted temporarily into
an emulsion by shaking, comprises at least one antimicrobial
active ingredient, together with a spray dispenser forms a
10 product and can be used in a method for the cleaning and/or
disinfection or sanitation of hard surfaces, in particular
glass, in which the liquid multiphase cleaner is converted
temporarily into an emulsion by shaking, is applied to the
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15 preferably by spraying, and this surface is then cleaned
and/or disinfected or sanitized, if necessary by wiping with
an absorbent soft article.

MULTIPHASE CLEANER WITH ANTIMICROBIAL ACTION**Field of the Invention**

5 The invention relates to multiphase liquid cleaners with antimicrobial action which can be emulsified temporarily by shaking, and which can be used for the cleaning of hard surfaces, in particular glass, to a product comprising the composition, and to a method for the cleaning and/or disinfection or sanitation of hard surfaces.

Background of the Invention

10 The hard-surface cleaners customary today are usually aqueous preparations in the form of a stable solution or dispersion which comprise, as essential active ingredients, surface-active substances, organic solvents and optionally
15 complexing agents for the hardness constituents of water, abrasive substances and alkalis which have a cleaning action. Cleaners intended primarily for the cleaning of glass and ceramic surfaces are often formulated as solutions
20 of the active ingredient in a mixture of water and water-miscible organic solvents, primarily lower alcohols and glycol ethers. Examples of such compositions are given in German laid-open application 22 20 540, US patent specifications 3 839 234 and 3 882 038, and in European
25 patent applications 344 847 and 393 772.

30 In the case of the use of cleaners, in addition to the requirement for high cleaning performance, as simple and convenient as possible an application of the compositions is also required. In most cases it is expected that the compositions produce the desired effect even after a single application without further measures. In this connection, especially in the case of application to smooth surfaces, in particular those like glass or ceramic which are able to
35 reflect in a sparkling manner, difficulties arise because compositions which clean well in most cases do not dry streak-free, while those compositions which dry essentially

without visible residues have only a limited cleaning action.

5 In order, for an acceptable residue behavior, to achieve a
sufficient cleaning action, in particular against greasy
soilings, it is necessary to also add relatively large
amounts of more or less volatile alkalis in addition to
organic solvents to the cleaners. In this regard, ammonia
and alkanolamines in particular have been used. However,
10 relatively high concentrations of ammonia or amines bring
about, as well as a notable odor nuisance, a corresponding
increase in the pH in the cleaner solution with the
consequence that relatively sensitive surfaces, such as, for
example, paint surfaces, are significantly attacked by these
15 cleaners.

There therefore continues to be a need for cleaners which,
for a high cleaning performance, do not have said
disadvantages.

20 Moreover, in addition to the high primary detergency of such
a cleaner, an antimicrobial action is nowadays also
increasingly expected. To this end, antimicrobial active
ingredients, for example surface-active quaternary
25 compounds, are incorporated into the cleaners.

German laid-open application 39 10 170 describes
compositions, used as mouthwashes, for bacterial desorption
from solid surfaces and living tissue, which are in the form
30 of a 2-phase preparation and which, upon shaking, form a
temporary oil-in-water emulsion of limited life span, the
aqueous phase comprising about 50 to 97% by weight and the
water-immiscible oil phase about 3 to 50% by weight. A
content of about 0.003 to 2% by weight of an amphiphilic
35 cationic agent, for example a cationic surfactant, is
essential, in an amount which permits the formation of the
oil-in-water emulsion, this emulsion cracking and separating

about 10 seconds to 30 minutes after its formation. Anionic surfactants impair the antibacterial action. Other surfactants are not mentioned.

5 European patent application 0 195 336 describes care compositions, which can be emulsified by shaking, for sensitive surfaces, in particular plastic surfaces or compact disks which, in addition to an aqueous phase, also
10 comprise an organic phase which consists of the chlorofluorocarbon (CFC) 1,1,2-trichloro-1,2,2-trifluoroethane. In addition, the compositions are free from wax. However, because of their environment-damaging properties, in particular in connection with the ozone hole, the use of CFCs should be reduced or, better still, avoided.

15 German laid-open applications 195 01 184, 195 01 187 and 195 01 188 (Henkel KGaA) disclose hair-treatment compositions in the form of a 2-phase system, which have an oil phase and a water phase, the oil phase preferably being
20 based on silicone oil, and which are temporarily miscible by mechanical action.

Summary of the Invention

The object of the present invention was then to provide
25 high-performance, storage-stable, easy-to-handle and antimicrobially effective compositions for the cleaning of hard surfaces.

In a first embodiment, the invention provides a liquid
30 multiphase cleaner having at least two continuous phases, which has at least one aqueous phase I and a nonaqueous liquid phase II which is immiscible with this aqueous phase, and which can be converted temporarily into an emulsion by shaking, and which comprises at least one antimicrobial
35 active ingredient.

For the purposes of the present invention, *immiscible, nonaqueous* phase means a phase *not based on water as solvent*, where, in the nonaqueous phase II, a certain amount, based on phase II, of water of customarily no more than 20% by weight, preferably no more than 15% by weight, in particular no more than 10% by weight, particularly preferably no more than 5% by weight, may be present in completely dissolved form.

In the simplest case, a composition according to the invention consists of a continuous aqueous phase which consists of the entire phase I, and a continuous nonaqueous liquid phase which consists of the entire phase II. One or more continuous phases of a composition according to the invention can, however, also comprise parts of another phase in emulsified form, meaning that, in such a composition, for example, part of phase I is in the form of continuous phase I, which represents the continuous aqueous phase of the composition, and another part is emulsified as discontinuous phase I in the continuous nonaqueous phase II. Analogous statements apply for phase II and further continuous phases.

In a second embodiment, the invention provides a method for the cleaning and/or disinfection or sanitation of hard surfaces, in which a cleaner according to the invention is converted temporarily into an emulsion by shaking, applied to the surface to be cleaned and/or disinfected or sanitized, preferably by spraying, and this surface is then cleaned and/or disinfected or sanitized, if necessary by wiping with an absorbent soft article.

For the purposes of the teaching according to the invention, the terms disinfection, sanitation, antimicrobial action and antimicrobial active ingredient have the customary specialist meanings given, for example, by K.H. Wallhäußer in "Praxis der Sterilisation, Desinfektion - Konservierung: Keimidentifizierung - Betriebshygiene" [Practice of

Sterilization, Disinfection - Preservation: Germ Identification - Plant Hygiene] (5th edition - Stuttgart; New York: Thieme, 1995). While disinfection in the narrower sense of medical practice means the killing of, in theory all, infection germs, sanitation means the elimination as far as possible of all germs, including saprophytic ones which are normally harmless for humans. In this connection, the extent of disinfection or sanitation is dependent on the antimicrobial action of the composition used, which decreases with decreasing content of antimicrobial active ingredient or increasing dilution of the composition for use.

The compositions according to the invention exhibit separate phases which can be emulsified for use, remain homogeneous during use and then again form separate phases, and have an antimicrobial action. Furthermore, as well as their high detergency, the compositions according to the invention are notable for an overall high storage stability. Thus, the individual phases in the composition are stable over a long period without the formation of, for example, deposits, and conversion to a temporary emulsion remains reversible even after frequent shaking. In addition, the physical form of the compositions according to the invention avoids per se the problem of stabilization of a composition formulated as an emulsion. Separation of ingredients into separate phases can additionally promote the chemical stability of the composition. In addition, the compositions according to the invention exhibit excellent residue behavior. Greasy residues are largely avoided, meaning that the surface shine is retained without rinsing being necessary.

Detailed Description of the Invention

The content of at least one antimicrobial active ingredient in the composition according to the invention is usually 0.001 to 20% by weight, preferably 0.01 to 10% by weight, in particular 0.02 to 5% by weight, particularly preferably

0.05 to 1.5% by weight and most preferably 0.1 to 0.5% by weight.

The composition according to the invention comprises, for the antimicrobial action, at least one antimicrobial active ingredient, preferably chosen from the group of alcohols, amines, aldehydes, antimicrobial acids or salts thereof, carboxylic esters, acid amides, phenols, phenol derivatives, diphenyls, diphenylalkanes, urea derivatives, oxygen-, nitrogen-acetals and formals, benzamidines, isothiazolines, phthalimide derivatives, pyridine derivatives, antimicrobial surface-active compounds, guanidines, antimicrobial amphoteric compounds, quinolines, 1,2-dibromo-2,4-dicyanobutane, iodo-2-propynylbutyl carbamate, iodine, iodophores and peroxo compounds, halogen compounds and any mixtures of the above.

Here, the antimicrobial active ingredient can be chosen from ethanol, n-propanol, isopropanol, 1,3-butanediol, phenoxyethanol, 1,2-propylene glycol, glycerol, undecylenic acid, citric acid, lactic acid, benzoic acid, salicylic acid, dihydracetic acid, o-phenylphenol, N-methylmorpholinium acetonitrile (MMA), 2-benzyl-4-chlorophenol, 2,2'-methylenebis(6-bromo-4-chlorophenol), 4,4'-dichloro-2'-hydroxydiphenyl ether (dichlosan), 2,4,4'-trichloro-2'-hydroxydiphenyl ether (trichlosan), chlorhexidine, N-(4-chlorophenyl)-N-(3,4-dichlorophenyl)urea, N,N'-(1,10-decanediyl)-1-pyridinyl-4-ylidene)bis(1-octanamine) dihydrochloride, N,N'-bis-(4-chlorophenyl)-3,12-diimino-2,4,11,13-tetraazatetradecanediimide amide, glucoprotamines, antimicrobial surface-active quaternary compounds, guanidines, including the bi- and polyguanidines, such as, for example, 1,6-bis(2-ethylhexylbiguanido)hexane dihydrochloride, 1,6-di(N₁,N₁'-phenyldiguanido-N₅,N₅')hexane tetrahydrochloride, 1,6-di(N₁,N₁'-phenyl-N₁,N₁'-methyldiguanido-N₅,N₅')hexane dihydrochloride, 1,6-di(N₁,N₁'-

o-chlorophenyldiguanido- N_5, N_5') hexane dihydrochloride,
 1,6-di (N_1, N_1' -2,6-dichlorophenyldiguanido- N_5, N_5') hexane
 dihydrochloride, 1,6-di [N_1, N_1' - β -(p-methoxyphenyl) diguanido-
 N_5, N_5'] hexane dihydrochloride, 1,6-di (N_1, N_1' - α -methyl- β -
 5 phenyldiguanido- N_5, N_5') hexane dihydrochloride, 1,6-di (N_1, N_1' -
 p-nitrophenyldiguanido- N_5, N_5') hexane dihydrochloride, $\omega:\omega$ -
 di (N_1, N_1' -phenyldiguanido- N_5, N_5')-di-n-propyl ether
 dihydrochloride, $\omega:\omega'$ -di (N_1, N_1' -p-chlorophenyldiguanido-
 N_5, N_5')-di-n-propyl ether tetrahydrochloride, 1,6-di (N_1, N_1' -
 10 2,4-dichlorophenyldiguanido- N_5, N_5') hexane
 tetrahydrochloride, 1,6-di (N_1, N_1' -p-methylphenyldiguanido-
 N_5, N_5') hexane dihydrochloride, 1,6-di (N_1, N_1' -
 2,4,5-trichlorophenyldiguanido- N_5, N_5') hexane tetra-
 hydrochloride, 1,6-di [N_1, N_1' - α -(p-chlorophenyl) ethyldi-
 15 guanido- N_5, N_5'] hexane dihydrochloride, $\omega:\omega'$ -di (N_1, N_1' -p-
 chlorophenyldiguanido- N_5, N_5')-m-xylene dihydrochloride,
 1,12-di (N_1, N_1' -p-chlorophenyldiguanido- N_5, N_5') dodecane
 dihydrochloride, 1,10-di (N_1, N_1' -phenyldiguanido- N_5, N_5') decane
 tetrahydrochloride, 1,12-di (N_1, N_1' -phenyldiguanido-
 20 N_5, N_5') dodecane tetrahydrochloride, 1,6-di (N_1, N_1' -o-
 chlorophenyldiguanido- N_5, N_5') hexane dihydrochloride, 1,6-di-
 (N_1, N_1' -o-chlorophenyldiguanido- N_5, N_5') hexane
 tetrahydrochloride, ethylenebis(1-tolylbiguanide),
 ethylenebis(p-tolylbiguanide), ethylene-
 25 bis(3,5-dimethylphenylbiguanide), ethylenebis(p-tert-
 amylphenylbiguanide), ethylenebis(nonylphenylbiguanide),
 ethylenebis(phenylbiguanide), ethylenebis(N-
 butylphenylbiguanide), ethylenebis(2,5-diethoxyphenyl-
 biguanide), ethylenebis(2,4-dimethylphenylbiguanide),
 30 ethylenebis(o-diphenylbiguanide), ethylenebis(mixed amyl-
 naphthylbiguanide), N-butylethylenebis(phenylbiguanide),
 trimethylenebis(o-tolylbiguanide), N-butyl-
 trimethylenebis(phenylbiguanide) and the corresponding
 salts, such as acetates, gluconates, hydrochlorides,
 35 hydrobromides, citrates, bisulfites, fluorides,
 polymaleates, N-cocoalkyl sarcosinates, phosphites,
 hypophosphites, perfluorooctanoates, silicates, sorbates,

salicylates, maleates, tartrates, fumarates, ethylenediaminetetraacetates, iminodiacetates, cinnamates, thiocyanates, arginates, pyromellitates, tetracarboxybutyrates, benzoates, glutarates, mono-
5 fluorophosphates, perfluoropropionates and any mixtures thereof. Also suitable are halogenated xylol and cresol derivatives, such as p-chlorometacresol or p-chlorometaxylol, amphoteric agents, and natural antimicrobial active ingredients of vegetable origin (e.g. from spices or herbs)
10 or of animal origin, and microbial origin. Preference may be given to using antimicrobially active surface-active quaternary compounds, a natural antimicrobial active ingredient of vegetable origin and/or a natural antimicrobial active ingredient of animal origin, most
15 preferably at least one natural antimicrobial active ingredient of vegetable origin from the group which includes caffeine, theobromine and theophylline and essential oils such as eugenol, thymol and geraniol, and/or at least one natural antimicrobial active ingredient of animal origin
20 from the group which includes enzymes such as milk protein, lysosyme and lactoperoxidase, and/or at least one antimicrobially active surface-active quaternary compound having an ammonium, sulfonium, phosphonium, iodonium or arsonium group, peroxo compounds and chlorine compounds.
25 Substances of microbial origin, so called bacteriocines, can also be used.

In a preferred embodiment, the composition according to the invention comprises at least one quaternary ammonium
30 compound (QAC) with antimicrobial action according to the general formula $(R^1)(R^2)(R^3)(R^4) N^+ X^-$, in which R^1 to R^4 are identical or different C_1 - C_{22} -alkyl radicals, C_7 - C_{28} -aralkyl radicals or heterocyclic radicals, where two, or in the case of an aromatic incorporation as in pyridine, even three
35 radicals, together with the nitrogen atom, form the heterocycle, e.g. a pyridinium or imidazolinium compound, and X^- are halide ions, sulfate ions, hydroxide ions or

similar anions. For optimal antimicrobial action, preferably at least one of the radicals has a chain length of from 8 to 18, in particular 12 to 16, carbon atoms.

5 QACs can be prepared by reacting tertiary amines with
alkylating agents, such as, for example, methyl chloride,
benzyl chloride, dimethyl sulfate, dodecyl bromide, and also
ethylene oxide. The alkylation of tertiary amines having one
10 long alkyl radical and two methyl groups proceeds
particularly readily, and the quaternization of tertiary
amines having two long radicals and one methyl group can
also be carried out using methyl chloride under mild
conditions. Amines which have three long alkyl radicals or
hydroxysubstituted alkyl radicals are less reactive and are
15 preferably quaternized using dimethyl sulfate.

Examples of suitable QACs are benzalkonium chloride
(N-alkyl-N,N-dimethylbenzylammonium chloride, CAS No. 8001-
54-5), benzalkone B (*m,p*-dichlorobenzyltrimethyl-C₁₂-
20 alkylammonium chloride, CAS No. 58390-78-6), benzoxonium
chloride (benzyl dodecylbis(2-hydroxyethyl)ammonium
chloride), cetrimonium bromide (N-hexadecyl-N,N-
trimethylammonium bromide, CAS No. 57-09-0), benzetonium
chloride (N,N-dimethyl-N-[2[2-[*p*-
25 (1,1,3,3-tetramethylbutyl)phenoxy]ethoxy]ethyl]benzyl-
ammonium chloride, CAS No. 121-54-0), dialkyl-
dimethylammonium chlorides such as di-*n*-decyl-
dimethylammonium chloride (CAS No. 7173-51-5-5),
didecyl dimethylammonium bromide (CAS No. 2390-68-3),
30 dioctyl dimethylammonium chloride, 1-cetylpyridinium chloride
(CAS No. 123-03-5) and thiazoline iodide (CAS No. 15764-48-
1) and mixtures thereof. Particularly preferred QACs are the
benzalkonium chlorides having C₈-C₁₈-alkyl radicals, in
particular C₁₂-C₁₄-alkylbenzyl dimethylammonium chloride.
35 Benzalkonium halides and/or substituted benzalkonium halides
are, for example, commercially available as Barquat[®] ex
Lonza, Marquat[®] ex Mason, Variquat[®] ex Witco/Sherex and

Hyamine[®] ex Lonza, and Bardac[®] ex Lonza. Further commercially available antimicrobial active ingredients are N-(3-chloroallyl)hexaminium chloride such as Dowicide[®] and Dowicil[®] ex Dow, benzethonium chloride such as Hyamine[®] 1622 ex Rohm & Haas, methylbenzethonium chloride such as Hyamine[®] 10X ex Rohn & Haas, cetylpyridinium chloride such as cepacol chloride ex Merrell Labs.

In a particular embodiment of the invention, the composition comprises two or more antimicrobial active ingredients, in particular at least one water-soluble antimicrobial active ingredient in phase I, for example a surface-active quaternary compound such as benzalkonium chloride and/or an antimicrobial acid such as benzoic acid or salicylic acid, and at least one further antimicrobial active ingredient in phase II, for example an essential oil such as eugenol, thymol or geraniol.

In a further preferred embodiment of the invention, the composition is *essentially CFC-free*, i.e. the nonaqueous liquid phase II is not based on CFCs. In this connection, because of the negative environmental effects of these compounds, the compositions according to the invention preferably comprise no CFCs at all, where small amounts, based on the overall composition, of up to about 5% by weight can still be tolerated.

In a preferred embodiment of the invention, the continuous phases I and II are delimited from one another by a sharp interface.

In a particular embodiment of the invention, one or both of the continuous phases I and II comprise parts, preferably 0.1 to 40% by volume, in particular 0.2 to 35% by volume, particularly preferably 0.5 to 30% by volume, most preferably 1 to 20% by volume, based on the volume of the respective continuous phase, of the other phase in each case

as dispersant. Here, the continuous phase I or II is then reduced by the part by volume which is distributed as dispersant in the other phase in each case. Particular preference is given here to compositions in which phase I is emulsified in phase II in amounts of from 0.1 to 40% by volume, in particular 0.2 to 35% by volume, for example about 33% by volume, based on the volume of phase II.

In a further particular embodiment of the invention, in addition to the continuous phases I and II, part of the two phases is in the form of an emulsion of one of the two phases in the other phase, this emulsion being delimited by two, preferably sharp, interfaces, one upper and one lower, from the parts of the phases I and II not involved in the emulsion.

The compositions according to the invention preferably comprise 5 to 95% by volume of phase I and 95 to 5% by volume of phase II.

In a further preferred embodiment of the invention, the composition comprises 35 to 95% by volume of phase I and 5 to 65% by volume of phase II, in particular 55 to 95% by volume of phase I and 5 to 45% by volume of phase II, most preferably 70 to 90% by volume of phase I and 10 to 30% by volume of phase II, for example 75, 80 or 85% by volume of phase I and 25, 20 or 15% by volume of phase II. In addition, the continuous phase I preferably represents the lower phase, and the continuous phase II the upper phase.

In a likewise preferred embodiment, the water-immiscible phase II is based on aliphatic petroleum hydrocarbons and/or terpene hydrocarbons. The petroleum hydrocarbons preferably have a boiling point range from 130 to 260°C, in particular from 140 to 240°C, particularly preferably from 150 to 220°C, such as, for example, the C₉₋₁₃-isoparaffins having a boiling point range from 184 to 217°C, available, for

example, as *Shellsol*[®] T from *Deutsche Shell Chemie AG* (Eschborn). Preferred terpene hydrocarbons are citrus oils such as the orange oil obtained from the skin of oranges, the orange terpenes present therein, in particular limonene, or pine oil, the tall oil extracted from roots and stumps, and mixtures thereof. Phase II can also consist entirely of aliphatic petroleum hydrocarbons and/or terpene hydrocarbons. In this connection, phase II comprises petroleum hydrocarbons preferably in amounts of from 60 to 100% by weight, particularly preferably 90 to 99.99% by weight, in particular 95 to 99.9% by weight, most preferably 97 to 99% by weight.

Suitable surface-active substances for the compositions according to the invention are surfactants, in particular from the classes of anionic and nonionic surfactants. The compositions preferably comprise one or more anionic and/or nonionic surfactants, where the anionic surfactants are in particular present in phase I. The amount of anionic surfactant, based on the phase I, is usually no more than 10% by weight, preferably between 0.01 and 5% by weight, in particular between 0.01 and 0.5% by weight, particularly preferably between 0.1 and 0.3% by weight. If the compositions comprise nonionic surfactants, the concentration thereof preferably in phase I, based on phase I, is usually no greater than 3% by weight, preferably between 0.001 and 0.3% by weight and in particular between 0.001 and 0.1% by weight, and in phase II, based on phase II, is usually no greater than 5% by weight, preferably between 0.001 and 0.5% by weight and in particular between 0.001 and 0.2% by weight, particularly preferably between 0.005 and 0.1% by weight, most preferably between 0.01 and 0.05% by weight.

Suitable as one or more anionic surfactants are preferably C₈-C₁₈-alkylbenzenesulfonates, in particular having about 12 carbon atoms in the alkyl moiety, C₈-C₂₀-alkanesulfonates,

C₈-C₁₈-monoalkyl sulfates, C₈-C₁₈-alkyl polyglycol ether sulfates having 2 to 6 ethylene oxide units (EO) in the ether moiety and sulfosuccinates having 8 to 18 carbon atoms in the alcohol radicals, and mixtures thereof.

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The anionic surfactants are preferably used as sodium salts, but can also be present as other alkali metal or alkaline earth metal salts, for example magnesium salts, and in the form of ammonium or amine salts.

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Examples of such surfactants are sodium cocoalkyl sulfate, sodium sec-alkanesulfonate having about 15 carbon atoms and sodium dioctyl sulfosuccinate. Fatty alkyl sulfates having 12 to 14 carbon atoms, and also sodium lauryl ether sulfate having 2 EO have proven particularly suitable.

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However, in accordance with the above quantitative data, if cationic antimicrobial active ingredients, such as surface-active quaternary compounds, in particular quaternary ammonium compounds, are used, the compositions preferably comprise as low as possible a content of anionic surfactant including soaps and, in a particularly preferred embodiment of the invention, are entirely free from anionic surfactants including soaps, since the latter are frequently more or less incompatible with the antimicrobially active quaternary compounds. In this connection, it is self-evident for the person skilled in the art that he or she must verify the compatibility of the anionic surfactants with the antimicrobial active ingredients with regard to the germ-reducing action.

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Nonionic surfactants which may be mentioned are especially C₈-C₁₈-alcohol polyglycol ethers, i.e. ethoxylated alcohols having 8 to 18 carbon atoms in the alkyl moiety and 2 to 15 ethylene oxide units (EO), C₈-C₁₈-carboxylic polyglycol esters having 2 to 15 EO, ethoxylated fatty acid amides having 12 to 18 carbon atoms in the fatty acid moiety and 2

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to 8 EO, long-chain amine oxides having 14 to 20 carbon atoms and long-chain alkyl polyglycosides having 8 to 14 carbon atoms in the alkyl moiety and 1 to 3 glycoside units. The compositions preferably comprise one or more nonionic surfactants from the group of ethoxylates of longer-chain alcohols, alkyl polyglycosides and mixtures thereof. Examples of such surfactants are oleyl cetyl alcohol having 5 EO, nonylphenol having 10 EO, lauric acid dyethanolamide, cocoalkyldimethylamine oxide and cocoalkyl polyglucoside having, on average, 1.4 glucose units.

Preferred nonionic surfactants in the aqueous phase are, as well as the addition products of ethylene oxide and longer-chain alcohols, in particular fatty alcohols, having in particular 4 to 8 ethylene oxide units, the alkyl polyglycosides, and of these in turn the representatives having 8 to 10 carbon atoms in the alkyl moiety and up to 2 glucose units. In the nonaqueous phase II particularly preferred nonionic surfactants are fatty alcohol polyglycol ethers having in particular 2 to 8 EO, for example oleyl cetyl alcohol + 5 EO ether, and/or fatty acid polyglycol esters (FAE) having in particular 2 to 10 EO, for example tallow fatty acid + 6 EO ester. Furthermore, in the case of the nonionic surfactants, in particular the alcohol polyglycol ethers and carboxylic polyglycol esters, for phase II, the degree of ethoxylation is matched to the carbon chain length such that relatively short carbon chains having lower degrees of ethoxylation, or relatively long carbon chains having relatively high degrees of ethoxylation are combined.

In a particular embodiment of the invention, the compositions comprise one or more anionic and nonionic surfactants. In this connection, combinations of anionic surfactant in phase I and nonionic surfactant in phase II are particularly advantageous, for example combinations of fatty alcohol sulfates and/or fatty alcohol polyglycol ether

sulfates in phase I with fatty alcohol polyglycol ethers and/or FAEs in phase II, in particular C₁₂-C₁₄-fatty alcohol sulfates and/or C₁₂-C₁₄-fatty alcohol polyglycol ether sulfates having, preferably, 2 ethylene oxide units (EO) in phase I and nonionic surfactant, in particular C₁₆-C₁₈-fatty alcohol polyglycol ethers having, preferably, 2 to 8 EO and/or C₁₄-C₁₈-fatty acid polyglycol esters having, preferably, 2 to 10 EO, in phase II.

Furthermore, the cleaners according to the invention can comprise one or more water-soluble organic solvents. The amount of one or more organic solvents in phase I is preferably 0.1 to 15% by weight, in particular 1 to 10% by weight, particularly preferably 2 to 8.5% by weight, most preferably 4 to 7% by weight, for example about 5 or 6% by weight, based on the aqueous phase I.

Examples of suitable solvents are water-soluble hydrocarbons having one or more hydroxyl groups, preferably the alcohols and glycols having 2 to 6 carbon atoms, in particular 2 or 3 carbon atoms, and ethers thereof in the form of the glycol ethers or ether alcohols, and mixtures thereof.

The alcohols used are in particular ethanol, isopropanol and n-propanol. Suitable ether alcohols are sufficiently water-soluble compounds having, preferably, no more than 10 carbon atoms in the molecule. Examples of such ether alcohols are ethylene glycol monobutyl ether, propylene glycol monobutyl ether, dyethylene glycol monobutyl ether, propylene glycol monotertiary-butyl ether and propylene glycol monoethyl ether, of which, in turn, ethylene glycol monobutyl ether and propylene glycol monobutyl ether are preferred. If alcohol and ether alcohol are used alongside one another, the weight ratio of the two is preferably between 1:2 and 4:1. Ethanol is particularly preferred for the purposes of the invention.

In a preferred embodiment, the composition according to the invention comprises one or more water-soluble organic solvents from the group of lower alcohols having 2 or 3 carbon atoms, of glycols having 2 or 3 carbon atoms and of glycol ethers having up to 10 carbon atoms, and mixtures thereof, in particular ethanol.

The compositions can be converted into the emulsion temporary in accordance with the invention by shaking preferably up to three times, in particular up to twice, particularly preferably once, the temporary emulsion generated by shaking being stable over a period sufficient for convenient application of the composition of from about 0.5 to 10 min, preferably 1 to 5 min, in particular 1.5 to 4 min, i.e. on the one hand does not crack again directly after shaking has finished, and on the other hand does not persist over a relatively long period. In this connection, *stable* means that after the respective time, at least 90% by volume of the composition is still in the form of the temporary emulsion generated by shaking. As well as the selection and concentration of the base components and active components, a rule for adjusting the ultimate properties of the compositions according to the invention consists in controlling the viscosity of the individual phases.

The aqueous phase I preferably has a viscosity according to *Brookfield* (model DV-II+, spindle 31, rate of rotation 20 min^{-1} , 20°C) of 0.1 to 200 mPa·s, in particular 0.5 to 100 mPa·s, particularly preferably 1 to 60 mPa·s and most preferably 5 to 20 mPa·s. For this purpose, the composition or the phases present may comprise one or more viscosity regulators. The amount of viscosity regulator in phase I, based on phase I, is usually up to 0.5% by weight, preferably 0.001 to 0.3% by weight, in particular 0.01 to 0.2% by weight, most preferably 0.02 to 0.1% by weight. Suitable viscosity regulators are inter alia synthetic

polymers such as the homo- and/or copolymers of acrylic acid or derivatives thereof, and salts thereof, for example the products available under the trade name Carbopol® from Goodrich, in particular the crosslinked acrylic acid copolymer Carbopol® EDT 2623. International application WO 97/38076, to which reference is made in this respect and the contents of which are hereby incorporated into this application, lists a number of other polymers derived from acrylic acid which are likewise suitable viscosity regulators.

Furthermore, the compositions according to the invention can comprise volatile alkali in phase I. As such alkali, ammonia and/or one or more alkanolamines which can contain up to 9 carbon atoms in the molecule are preferred. Preferred alkanolamines are the ethanolamines, and of these in turn monoethanolamine. The content of ammonia and/or alkanolamine is, based on phase I, preferably 0.01 to 3% by weight, in particular 0.02 to 1% by weight, particularly preferably 0.05 to 0.5% by weight.

In addition to the volatile alkali, the compositions according to the invention can additionally comprise carboxylic acid in phase I, the equivalent ratio of amine and/or ammonia to carboxylic acid preferably being between 1:0.9 and 1:0.1. Suitable carboxylic acids have up to 6 carbon atoms and are mono-, di- or polycarboxylic acids. Depending on the equivalent weight of amine and carboxylic acid, the content of carboxylic acid, based on phase I, is preferably between 0.01 and 2.7% by weight, in particular between 0.01 and 0.9% by weight. Examples of suitable carboxylic acids are acetic acid, glycolic acid, lactic acid, citric acid, succinic acid and adipic acid, of which preference is given to using acetic acid, citric acid and lactic acid. Particular preference is given to using acetic acid.

In addition to said components, the compositions according to the invention can comprise further auxiliaries and additives as are customary in such compositions. These include, in particular, dyes, perfume oils, pH regulators (e.g. citric acid, alkanolamines or NaOH), preservatives, 5 complexing agents for alkaline earth metal ions, enzymes, bleach systems and antistats. The amount of such additives is usually no more than 2% by weight in the cleaner. The lower use limit depends on the nature of the additive and, 10 for example in the case of dyes, can be up to 0.001% by weight and below. The amount of auxiliaries is preferably between 0.01 and 1% by weight.

The pH of the aqueous phase I can be varied over a wide 15 range from strongly acidic via neutral to highly alkaline, but is preferably in a range from 1 to 12, in particular 2 to 11. In this connection, the pH dependency - either known to the person skilled in the art or accessible by routine experiments - of the antimicrobial action of the 20 antimicrobial active ingredient(s) used is to be taken into consideration. Thus, the pH is to be chosen accordingly to be as optimal as possible for the antimicrobial action of the respective antimicrobial active ingredient(s) and/or if a certain pH is prescribed for a certain cleaning purpose of 25 the antimicrobial active ingredient(s).

In a neutral variant, the pH is greater than 6 to less than 8, preferably 6.5 to 7.5 and in particular about 7.

30 In a preferred embodiment of the invention, the compositions are rendered alkaline with a pH of from 8 to 12, preferably 8 to 11, in particular 8 to 10.5, for example between 8 and 9 for moderate alkalinity or greater than 9 to 10.5, 11 or even 12, e.g. 10, for stronger alkalinity. This is preferred 35 particularly if antimicrobial quaternary compounds are used, since the antimicrobial action thereof usually increases

with increasing pH, while it is sometimes completely absent in the acid range below a pH of 3.

5 In a likewise preferred embodiment of the invention, the compositions are rendered acidic with a pH of from 1 to 6, preferably 1.5 to 5.5, in particular 2 to 5, particularly preferably 2.2 to 4.5, for example 2.3, 3 or 4. This is preferable, for example, if salicylic acid is used as the antimicrobial active ingredient since the antimicrobial
10 action thereof usually increases with decreasing pH.

In a preferred embodiment, the compositions according to the invention comprise 70 to 95% by volume, in particular 70 to 90% by volume, of aqueous phase I comprising

15 0 to 10% by weight of anionic surfactant,
0 to 3% by weight of nonionic surfactant,
0 to 2% by weight of at least one antimicrobial active ingredient,

0 to 10% by weight of water-soluble organic solvent,

20 0 to 0.5% by weight of viscosity regulator,

0 to 3% by weight of volatile alkali,

0 to 0.2% by weight of perfume and

ad 100% by weight of water, and

25 5 to 30% by volume, in particular 10 to 30% by volume, of nonaqueous phase II comprising

0 to 100% by weight of aliphatic petroleum hydrocarbons,

0 to 100% by weight of terpene hydrocarbons,

0 to 10% by weight of at least one antimicrobial active ingredient,

30 0 to 5% by weight of nonionic surfactant and

0 to 1% by weight of perfume,

where the % by weight figures are based on the respective phase, the sum of the petroleum and terpene hydrocarbons makes up phase II ad 100% by weight, at least one of the two
35 phases I and II comprises at least one antimicrobial active ingredient in an amount, based on the respective phase, of more than 0% by weight, and the phases may additionally

comprise in particular small amounts of dye. Particularly preferably, the anionic surfactants are present in amounts of from 0.01 to 5% by weight, preferably 0.01 to 0.5% by weight, in particular 0.1 to 0.3% by weight, and/or the
5 nonionic surfactants are present in amounts of from 0.001 to 0.5% by weight, preferably 0.005 to 0.1% by weight, in particular 0.01 to 0.05% by weight.

10 The composition according to the invention is sprayable and can therefore be used in a spray dispenser.

Thirdly, the invention accordingly provides a product comprising a composition according to the invention and a
15 spray dispenser.

The spray dispenser is preferably a manually activated spray dispenser, in particular chosen from the group comprising aerosol spray dispensers, self-pressurizing spray
20 dispensers, pump spray dispensers and trigger spray dispensers, in particular pump spray dispensers and trigger spray dispensers having a container made of transparent polyethylene or polyethylene terephthalate. Spray dispensers are described in more detail in WO 96/04940 (*Procter & Gamble*) and the US patents cited therein relating to spray
25 dispensers, to all of which reference is made in this regard and the contents of which are hereby incorporated into this application.

30 The compositions according to the invention are preferably used by applying the composition converted temporarily into an emulsion by shaking in amounts of about 1.5 to 10 g per m², in particular 3 to 7 g per m², to the surface to be cleaned and/or disinfected or sanitized, and directly
35 thereafter wiping this surface with an absorbent soft article and cleaning and/or disinfecting or sanitizing it in the process. The compositions are preferably applied using suitable spray devices, in particular a spray dispenser or a

product according to the invention in order to achieve as uniform a distribution as possible. Sponges or cloths in particular are suitable for the wiping, which, in the case of treatment of relatively large surfaces, can be rinsed out
5 with water from time to time.

The compositions according to the invention can be prepared by separately mixing the individual phases directly from their raw materials, subsequently combining and thoroughly
10 mixing the phases and finally allowing the composition to stand for separation of the temporary emulsion. They can also be prepared by mixing directly from their raw materials, subsequently thoroughly mixing and finally
15 allowing the composition to stand for separation of the temporary emulsion. If one component is not entirely insoluble in a phase other than that to which the respective component has been assigned or with which it has been
incorporated into the composition, this other phase may also
20 comprise corresponding proportions of the component in question within the framework of the establishment of solubility equilibria as a result of diffusion.

Examples

5 The makeup of the compositions E1 and E2 according to the invention, given by way of example, is given in Table 1. The aliphatic petroleum hydrocarbon used was C₉₋₁₃-isoparaffins. The compositions comprised a transparent aqueous phase I at the bottom and a creamy emulsion of phase I in nonaqueous phase II at the top, the volume of emulsified phase I in each case constituting about one third of the volume of phase II, in a volume ratio of phase I to the emulsion of phase I in phase II of about 80 to 20 and separated by a sharp interface. The temporary emulsion generated by shaking remained stable over a period of about 3 min, sufficient for the easy application of the composition, after which the phases separated again.

Table 1

Composition [% by weight]	E1	E2
C ₁₂₋₁₄ -alkylbenzyltrimethylammonium chloride	0.02	-
Salicylic acid	-	0.9
C ₈₋₁₀ -alkyl 1.5 glucoside	0.2	-
Sodium C ₁₂₋₁₄ fatty alcohol sulfate	-	0.2
Ethanol	4.8	4.8
Crosslinked acrylic acid copolymer ^[a]	0.025	0.025
Aliphatic petroleum hydrocarbon ^[b]	15	15
Aqueous ammonia solution, 25% strength by weight	0.046	0.046
Perfume	0.06	0.06
Dye	< 0.001	< 0.001
Water	ad 100	ad 100
pH of phase I	10	2.3

^[a] Carbopol® ETD 2623

^[b] Shellsol® T

Patent Claims:

1. A liquid multiphase cleaner composition having at least two continuous phases comprising at least one aqueous phase and a nonaqueous liquid phase which is immiscible with the aqueous phase, the cleaner composition being capable of conversion temporarily into an emulsion by shaking, and having at least one antimicrobial active ingredient.
2. The composition as claimed in claim 1, comprising at least one antimicrobial active ingredient in an amount of from 0.001 to 20% by weight.
3. The composition as claimed in claim 2, wherein the amount is 0.01 to 10% by weight.
4. The composition as claimed in claim 2, wherein the amount is 0.02 to 5% by weight.
5. The composition as claimed in claim 2, wherein the amount is 0.05 to 1.5% by weight.
6. The composition as claimed in claim 2, wherein the amount is 0.1 to 0.5% by weight.
7. The composition as claimed in any of claims 1 to 6, comprising at least one natural antimicrobial active ingredient of vegetable origin selected from the group comprising caffeine, theobromine and theophylline and essential oils selected from eugenol, thymol and geraniol and/or at least one natural antimicrobial active ingredient of animal origin.
8. The composition as claimed in claim 7, wherein the natural antimicrobial active ingredient of animal

origin which includes enzymes is selected from milk protein, lysosyme and lactoperoxidase.

9. The composition as claimed in any of claims 1 to 8,
which comprises at least one surface-active quaternary
compound having an ammonium, sulfonium, phosphonium,
iodonium or arsonium group.
10. The composition as claimed in claim 9, which comprises
at least one quaternary ammonium compound.
11. The composition as claimed in any of claims 1 to 10,
which comprises two or more antimicrobial active
ingredients.
12. The composition as claimed in claim 11, wherein at
least one water-soluble antimicrobial active ingredient
is present in the aqueous phase and at least one
further antimicrobial active ingredient is present in
the non-aqueous phase.
13. The composition as claimed in any of claims 1 to 12,
wherein the continuous phases are delimited from one
another by a sharp interface.
14. The composition as claimed in any of claims 1 to 13,
wherein one or both of the continuous phases comprise
parts based on the volume of the respective continuous
phase of the other phase as dispersant.
15. The composition as claimed in claim 14, wherein the
parts of the other phase as dispersant comprise 0.1 to
40% by volume based on the volume of the respective
continuous phase.
16. The composition as claimed in claim 14, wherein the
parts of the other phase as dispersant comprise 0.2 to

35% by volume based on the volume of the respective continuous phase.

- 5 17. The composition as claimed in claim 14, wherein the parts of the other phase as dispersant comprise 0.5 to 30% by volume based on the volume of the respective continuous phase.
- 10 18. The composition as claimed in claim 14, wherein the parts of the other phase as dispersant comprise 1 to 20% by volume based on the volume of the respective continuous phase.
- 15 19. The composition as claimed in any of claims 1 to 18, wherein the aqueous phase is emulsified in the non-aqueous phase in amounts of from 0.1 to 40% by volume, based on the volume of the non-aqueous phase.
- 20 20. The composition as claimed in claim 19, wherein the amounts are 0.2 to 35% by volume.
- 25 21. The composition as claimed in any of claims 1 to 20, wherein, in addition to the continuous phases, part of the two phases is in the form of an emulsion of one of the two phases in the other phase, this emulsion being delimited by two sharp interfaces, one upper and one lower, from the parts of the continuous phases not involved in the emulsion.
- 30 22. The composition as claimed in any of claims 1 to 21, which comprises 35 to 95% by volume of aqueous phase and 5 to 65% by volume of non-aqueous phase.
- 35 23. The composition as claimed in claim 22, wherein the composition comprises 55 to 95% by volume of aqueous phase and 5 to 45% by volume of non-aqueous phase.

24. The composition as claimed in claim 22, wherein the composition comprises 70 to 90% by volume of aqueous phase and 10 to 30% by volume of non-aqueous phase.
- 5 25. The composition as claimed in any of claims 1 to 24, which comprises a non-aqueous phase which consists of or is based on aliphatic petroleum hydrocarbons and/or terpene hydrocarbons.
- 10 26. The composition as claimed in claim 25, wherein the aliphatic petroleum hydrocarbons have a boiling range from 130 to 260°C and/or terpene hydrocarbons are orange oil and/or pine oil.
- 15 27. The composition as claimed in claim 25 or 26, wherein the boiling range is 140 to 240°C.
28. The composition as claimed in claim 25 or 26, wherein the boiling range is 150 to 220°C.
- 20 29. The composition as claimed in any of claims 1 to 28, which comprises, in the non-aqueous phase, aliphatic petroleum hydrocarbons in amounts, based on the non-aqueous phase, of from 60 to 100% by weight.
- 25 30. The composition as claimed in claim 29, wherein the amounts are 90 to 99.99% by weight.
31. The composition as claimed in claim 29, wherein the amounts are 95 to 99.9% by weight.
- 30 32. The composition as claimed in claim 29, wherein the amounts are 97 to 99% by weight.
- 35 33. The composition as claimed in any of claims 1 to 32, which comprises one or more anionic and/or nonionic surfactants.

34. The composition as claimed in any of claims 1 to 33,
which comprises one or more anionic surfactants
selected from the group comprising C₈-C₁₈-alkylbenzene-
sulfonates, C₈-C₂₀-alkanesulfonates, C₈-C₁₈-monoalkyl
5 sulfates, C₈-C₁₈-alkyl polyglycol ether sulfates having
2 to 6 ethylene oxide units, and C₈-C₁₈-alkyl alcohol
sulfosuccinates and mixtures thereof.
35. The composition as claimed in any of claims 1 to 34,
10 which comprises one or more nonionic surfactants from
the group comprising ethoxylates of longer-chain
alcohols, alkyl polyglycosides and mixtures thereof.
36. The composition as claimed in any of claims 1 to 35,
15 which comprises one or more anionic and nonionic
surfactants.
37. The composition as claimed in any of claims 1 to 36,
20 which comprises, in the aqueous phase, an anionic
surfactant selected from C₁₂-C₁₄-fatty alcohol sulfates
and/or C₁₂-C₁₄-fatty alcohol polyglycol ether sulfates
and in the non-aqueous phase, a nonionic surfactant
selected from C₁₆-C₁₈-fatty alcohol polyglycol ethers
and/or C₁₄-C₁₈-fatty acid polyglycol esters.
25
38. The composition as claimed in claim 37, wherein the
C₁₂-C₁₄-fatty alcohol polyglycol ether sulfates have 2
ethylene oxide units (EO), the C₁₆-C₁₈-fatty alcohol
polyglycol ethers have 2 to 8 EO, and the C₁₄-C₁₈-fatty
30 acid polyglycol esters have 2 to 10 EO.
39. The composition as claimed in any of claims 1 to 38,
which comprises one or more water-soluble organic
solvents.
35
40. The composition as claimed in claim 39, wherein the
organic solvents are selected from the group comprising

lower alcohols having 2 or 3 carbon atoms, glycols having 2 or 3 carbon atoms, glycol ethers having up to 10 carbon atoms, and mixtures thereof.

- 5 41. The composition as claimed in claim 39 or 40, wherein the organic solvent is ethanol.
- 10 42. The composition as claimed in any of claims 1 to 41, which comprises, in the aqueous phase, 0.1 to 15% by weight based on the aqueous phase, of solvent.
- 15 43. The composition as claimed in claim 42, wherein the solvent comprises 1 to 10% by weight of the aqueous phase.
- 20 44. The composition as claimed in any of claims 1 to 43, which comprises one or more viscosity regulators selected from homo- and/or copolymers of acrylic acid or derivatives thereof, and salts thereof, in amounts, based on the aqueous phase of up to 0.5% by weight.
- 25 45. The composition as claimed in claim 44, wherein the amounts are 0.001 to 0.3% by weight.
- 30 46. The composition as claimed in claim 44, wherein the amounts are 0.01 to 0.2% by weight.
- 35 47. The composition as claimed in claim 44, wherein the amounts are 0.02 to 0.1% by weight.
48. The composition as claimed in any of claims 1 to 47, which comprises an aqueous phase having a viscosity of from 0.1 to 200 mPa·s.
49. The composition as claimed in claim 48, wherein the viscosity of the aqueous phase is 0.5 to 100 mPa·s.

50. The composition as claimed in claim 48, wherein the viscosity of the aqueous phase is 1 to 60 mPa·s.
51. The composition as claimed in any of claims 1 to 50,
5 which comprises, in the aqueous phase, volatile alkali selected from ammonia and/or one or more alkanolamines having up to 9 carbon atoms.
52. The composition as claimed in claim 51, wherein the
10 volatile alkali is present in amounts, based on the aqueous phase, of from 0.01 to 3% by weight.
53. The composition as claimed in claim 51, wherein the
15 volatile alkali is present in amounts, based on the aqueous phase, of from 0.02 to 1% by weight.
54. The composition as claimed in claim 51, wherein the
20 volatile alkali is present in amounts, based on the aqueous phase, of from 0.05 to 0.5% by weight.
55. The composition as claimed in any of claims 1 to 54,
which comprises 70 to 95% by volume of the aqueous phase comprising
- 25 0 to 10% by weight of anionic surfactant,
0 to 3% by weight of nonionic surfactant,
0 to 2% by weight of at least one antimicrobial active ingredient,
0 to 10% by weight of water-soluble organic solvent,
- 30 0 to 0.5% by weight of viscosity regulator,
0 to 3% by weight of volatile alkali,
0 to 0.2% by weight of perfume and
ad 100% by weight of water, and
5 to 30% by volume of the non-aqueous phase
35 comprising
0 to 100% by weight of aliphatic petroleum hydrocarbons,

0 to 100% by weight of terpene hydrocarbons,
0 to 10% by weight of at least one antimicrobial
active ingredient,
0 to 5% by weight of nonionic surfactant and
0 to 1% by weight of perfume,

where the % by weight figures are based on the
respective phase, the sum of the aliphatic petroleum
and terpene hydrocarbons makes up phase II ad 100% by
weight and at least one of the two phases comprises at
least one antimicrobial active ingredient in an amount,
based on the respective phase, of more than 0% by
weight.

56. The composition as claimed in any of claims 1 to 55,
which comprises the anionic surfactants in amounts of
from 0.01 to 5% by weight and/or the nonionic
surfactants in amounts of from 0.001 to 0.5% by weight.

57. The composition as claimed in claim 56, wherein the
anionic surfactants comprise 0.01 to 0.5% by weight and
the nonionic surfactants comprise 0.005 to 0.1% by
weight.

58. The composition as claimed in claim 56, wherein the
anionic surfactants comprise 0.1 to 0.3% by weight and
the nonionic surfactants comprise 0.01 to 0.05% by
weight.

59. A product comprising a composition as claimed in any of
composition claims 1 to 58 in a spray dispenser.

60. A method for the cleaning and/or disinfection or
sanitation of hard surfaces in which a cleaner as
claimed in any of the above composition claims is
converted temporarily into an emulsion by shaking,
applied to the surface to be cleaned and/or disinfected
or sanitized, and this surface is then cleaned and/or

disinfected or sanitized, and if necessary, by wiping with an absorbent soft article.

5

61. A method as claimed in claim 60, wherein the surface is a glass surface.
62. A method as claimed in claim 60 or 61 wherein the composition is applied by spraying.